ATMOSPHERIC RADIO-ACTIVITY IN CALIFORNIA AND COLORADO AND THE RANGE OF THE α-PARTICLES FROM RADIUM B

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ATMOSPHERIC RADIOACTIVITY IN CALIFORNIA AND COLORADO AND THE RANGE OF THE α-PARTICLES FROM RADIUM B.[1]

BY FREDERIC A. HARVEY.

THIS paper is an account of an investigation of atmospheric radioactivity at Berkeley California, and Denver, Colorado. The work is of interest in itself, since no previous investigation of this character has been made in these localities; and several new results have also been obtained, including the proof that radium B gives off α-rays of very short range, and the measurement of this range.[2]

The earliest investigations on the subject of atmospheric radioactivity were made by Elster and Geitel.[3] They found that a charged body loses electricity faster in the open air than can be explained by leakage across its supports and that this was due to a slight ionization of the air, which always exists. In the light of the recent discoveries with regard to radioactivity it was seen that this ionization might be caused by some radioactive substance in the atmosphere. To test this they exposed a negatively charged wire gauze in the open air. After an exposure of a few hours the wire gauze was placed in the ionization chamber of an electroscope and it was found that an active deposit had collected on the gauze. The amount of active deposit collected was greatly increased by using a long wire and changing the negative potential from four hundred to several thousand volts. The material of the wire had no effect on the amount of active material collected. The activity decayed in a few hours in a manner resembling that of thorium X. The decay was not hastened by heating. The active deposit could

[1] Submitted in partial satisfaction of the requirements for the degree of Ph.D. in the University of California.

[2] A brief note containing some of the more important points has already been published by the author. Phys. Zeitschr., 1909.

[3] Elster and Geitel, Phys. Zeitschr., 3, p. 76, 1901, and p. 574, 1902; also 4, p. 522, 1903.

be transferred to a piece of leather and when concentrated in this form was sufficiently active to make a barium-platino-cyanide screen glow quite perceptibly to the rested eye, or give a shadow picture through a sheet of aluminium 0.1 mm. in thickness. All of these properties tended to show that the material was of the same nature as the active deposit collected from radium or thorium emanation. The amount of active material collected in caves and holes, where the air had been long stagnant, was much greater than in the open air. Meteorological conditions, such as fog, strong winds, temperature, and barometric height, were found to exert a marked effect on the amount of active material collected.

The work of Rutherford and Soddy[1] on the time rate of decay of thorium X has been extended until it is now generally accepted that each radioactive substance loses its activity according to some exponential law. Thus it is estimated that radium would lose half of its activity in about 1,760 years, whereas, radium A sinks to half-value in only three minutes. Where an active deposit consists of several radioactive components the total activity is made up of the sum of the activities due to these components. The usual method of analysis is to compare the curve showing the rate of decay of the active material with the curves of decay of the transformation products of radium, thorium and actinium, which are known with considerable accuracy. The method is more or less "cut and try" until a combination is found which will make the calculated and experimental results agree.

It is of importance to find the source of the emanation present in the air. Working toward this end Bumstead and Wheeler[2] found that the gas drawn from the surface soil at New Haven contained a radioactive gas apparently identical with radium emanation. Thorium emanation was not discovered because its life is shorter than the necessary time which elapsed in transferring the gas to the testing apparatus. Later Dadourian[3] exposed a negatively charged wire in a hole dug in the ground, at New Haven, into which fresh air was continually drawn from the surrounding soil. He found

[1] Rutherford and Soddy, Phil. Mag., September and November, 1902.

[2] Bumstead and Wheeler, Amer. Jour. Sc., Vol. 17, p. 97, February, 1904.

[3] H. M. Dadourian, Amer. Jour. Sc., Vol. 19, p. 16, January, 1905.

undoubted evidence of thorium emanation in the soil gases. Eve has shown that the amount of radium salts distributed through the earth's crust is of the right order of magnitude to account for the emanation in the atmosphere.

The nature of the emanation which produces the phenomena of atmospheric radioactivity has been the subject of investigation by many observers with apparently diverse results. The usual method is to observe the rate of decay of the active deposit collected on a negatively charged wire exposed in the open air and compare it with the known and different rates of decay of radium, thorium and actinium transformation products. Allen[1] working at Montreal and Cincinnati came first to the conclusion that the emanation is identical with that from radium, but later became convinced, owing to the diverse values of the half-period of decay (viz., the time taken for the decaying radioactive substance to lose half of its initial activity) of the active deposit, that it is a mixture, in varying proportions, of the emanations from radium, thorium and, possibly, actinium. Bumstead[2] showed that the active deposit collected on a wire exposed in the open air at New Haven is complex and that the thorium active deposit sometimes amounts to 15 per cent. of the total. Blanc[3] found that at Rome a very large percentage of the active deposit, from 50 to 70 per cent., collected on a wire exposed at a potential of — 500 volts, was due to thorium transformation products. The exposure in this case was long; nearly three days.

The objects of the present investigation are as follows: (1) To carry out for Berkeley, California, investigations similar to those which have been made in other localities as to the effect of meteorological conditions and as to the nature of the active deposit collected on a negatively charged wire exposed in the open air. (2) To determine the conditions governing the proportion of the components of the active deposit since this proportion was found to be variable. (3) To compare the active deposit obtained at Denver, Colorado, with that obtained at Berkeley. (4) To measure the range of the α-particles given off by the active deposit, particularly the range of

[1] S. J. Allen, Phil. Mag., December, 1904. PHYS. REV., June, 1908.

[2] H. A. Bumstead, Amer. Jour. Sc., Vol. 18, July, 1904.

[3] G. A. Blanc, Phil. Mag., 13, p. 378, March, 1907.

the very easily absorbed radiation and to determine to what component this radiation is due.

The usual method for collecting the radioactive material was used, viz., a wire charged to a negative potential of a few thousand volts, was exposed in the open air for several hours. The wire was of copper, No. 22, B. and S. gauge, about 14 meters in length, supported about four meters from the ground, in the earlier experiments. Later the length was increased to 38 meters, the distance from the ground to about 5 meters, and the wire stretched in a position where winds from all directions could reach it easily. For insulating supports thin ebonite rods, about 40 cm. in length, were used. It was found that in damp weather a thin coating of vaseline put on the hot rods would greatly improve the insulation, as a film of moisture does not collect as readily on the vaseline as on the ebonite. The potential of the wire was maintained at first by means of a water-dropper and later by means of a motor-driven Wimshurst machine, Tudsbury patent. The Wimshurst machine worked more satisfactorily in damp weather than the water-dropper. The potential was measured by means of a Braun electrometer reading directly in volts.

In order to analyze the active deposit it is first necessary to obtain observations which show its rate of decay. This decay, always measured by means of the ionization produced by the α-rays, was followed by several methods:

1. By means of an aluminium-leaf electroscope with sulphur insulation. The rate of fall of the leaf was measured in three ways. (*a*) The image of a circular scale, graduated in degrees, of radius equal to the length of the aluminium leaf, was superimposed on the leaf by means of a half-silvered mirror, placed at an angle of 45° in front of the observing telescope, and readings of the position of the leaf were taken at regular intervals. A curve was then plotted with positions of the leaf and times of observation as coördinates. The tangents to this curve give the rate of fall of the leaf. Any error in reading is self-correcting by this method and tangents can be determined within about one per cent. (*b*) A small Gurley comparator, reading to $\frac{1}{100}$ mm., was used. Readings were taken of the distance moved by the end of the leaf, at about 45° deflection,

during regular intervals of time. (*c*) This method differs from the first only in that a circular scale, with vernier, was fixed to the tube of a telescope and the angular motion of the leaf read by turning the cross wires until one of them coincided with the aluminium leaf. When a very large amount of active material had been collected sufficient accuracy could be attained, without plotting the curve of position, by merely observing the angular fall of the leaf during short, equal intervals of time, from one to ten minutes. The electroscope was calibrated by noting the rate of fall over various parts of the scale when a constant source of ionization, viz., the α-rays from uranium oxide, was used. The deflection of the leaf was kept between the limits 45° and 20°, as over this part of the scale the rate of fall is most uniform. The potential of the charged system for a deflection of 20° was about 500 volts, which was more than sufficient to secure the saturation current for the degree of ionization obtained.

2. By means of a quadrant electrometer made by Max Kohl after the design of Elster and Geitel, which was connected up in the usual way; one pair of quadrants earthed, the other pair connected to a plate or rod in the ionization chamber and the needle charged to a potential of 100 volts. The leak to the quadrants was then measured by noting, with a stop watch, the time taken for the spot of light to travel over a definite number of divisions on the scale — always the same divisions.

3. The null method described by S. J. Allen[1] was used. In this method the unknown ionization is balanced against a known ionization produced by a thin layer of uranium oxide. The parallel plates or concentric cylinders between which is the unknown ionization, are connected to one end of a battery of small cells while the uranium oxide plates are connected to the other end, the middle point being earthed in such a way that the current to one pair of quadrants from the unknown is of opposite sign to that from the standard. The surface of the uranium oxide exposed is varied until these two currents are just equal, when there is no change in the deflection of the needle. The instrument is calibrated for different amounts of surface exposed.

[1] S. J. Allen, Phil. Mag., December, 1907.

The first method was the least subject to external disturbing influences and was sufficiently accurate except when a comparison of the amount of activity of two substances was desired. In consequence it was most used. The other methods, however, were useful as checks. It is not possible to distinguish the curves obtained by one method from those obtained by another.

Percentages of thorium excited activity were determined in the following manner: Several sets of observations on the rate of decay of the active deposit from radium for a three-hour exposure of a negatively charged wire (potential — 5,000 volts or over) to radium emanation in the bell-jar were taken, using the ionization produced by the α-rays. These sets of observations were reduced to a common scale and averaged. From these average values a standard curve was plotted for the rate of decay of an equilibrium mixture of radium A, B and C. The agreement among the several sets of observations was good, the divergence being not more than two or three per cent. in any case. The standard curve was then made to agree with the curve under examination at the thirty-minute point and the activity due to the radium deposit calculated for the three-hour point. Using this value as a first approximation the difference between it and the experimental value gives the activity due to the thorium active deposit at the three-hour point. From this the thorium activity at the thirty-minute point was calculated, using the equation $I = I_0 e^{-\lambda t}$. Thorium active deposit loses half its activity in about eleven hours, hence $I = \frac{1}{2} I_0 = I_0 e^{-11\lambda}$ and $\lambda = .063$ when t, the time, is in hours. The part of the activity due to the radium deposit could now be determined for the thirty-minute point. Using this more accurate value the radium activity at the three-hour point was re-determined from the standard curve, and using the difference between this second approximation and the experimental value as the activity due to the thorium active deposit at the three-hour point the initial amount was determined. This divided by the total initial activity gave the percentage of thorium excited activity to a very close approximation. Correction for the normal leak of the electrometer or electroscope was of course made in each case. When observations had been carried on over a length of time as great as eleven hours the amount of thorium excited activity could be at

once determined, since usually all of the radium excited activity disappears within six hours from the time of ending the exposure. Calculations by the first method showed very good agreement with those by the second.

Observations Made in Berkeley.

The half-value period of the active deposit obtained on a negatively charged wire exposed in the open air was not constant, but varied between wide limits. In particular (see Table I.) the half-period varied from 33 minutes to 5½ hours. As will be shown later the active deposits can be accounted for on the supposition that they consist entirely of radium and thorium transformation products and this change in the period is due to the varying proportions of the components. It takes a little over three days for the thorium active deposit, which will collect on a negatively charged body to reach its maximum value, provided all the conditions are constant during this interval, which is impossible in an open air exposure, and the period will increase with the percentage of thorium deposit. Thus the period depends to a certain extent upon the time of exposure.

This variation of the half-period has also been observed by S. J. Allen.[1] A large part of the present investigation had been completed before Allen's work appeared. The agreement at the two localities is good except that the period varies between wider limits at Berkeley, an effect due to two causes. The present method allows that observations be commenced sooner after the discharge of the wire than does that of Allen. This makes it possible, at times, to observe the activity due to the very rapidly decaying radium A and consequently shortens the period observed. Greater percentages of thorium active deposit must account for the extension of the other limit.

The agreement between the calculated values, assuming that the excited activity is due to radium and thorium and the observed values, is very good. After the percentage of thorium active deposit initially on the wire had been calculated, according to the method described above, the curve $I = I_0 e^{-\lambda t}$ was plotted, where

[1] S. J. Allen, Phys. Rev., Vol. 26, p. 483, June, 1908.

TABLE I.

No. of Curve.	Half-Period.	Per cent. of Th Initially.	Time of Exposure.	Potential.	Remarks.
1	44 min.	1.8	3½ hrs.	3,300	Clear, light east wind, dry. Barom. steady, nearly logarithmic.
2	47 "	3.0	4 "	3,300	ditto.
3	43 "	4.0	3 "	3,600	North wind, clear, hot, very dry, nearly logarithmic.
4	34 "	6.0	3 "	5,000	North wind.
5	72 "	12.8	12¾ "	4,000	North wind, over-night exposure, with strong wind.
6	58 "	12.8	14½ "	4,000	ditto.
7	48 "	22.0	3 "	3,800	Light west wind.
8	66 "	19.0	4 "	3,250	Light southwest wind, very clear, absolutely logarithmic.
9	89 "	46.0	5½ "	3,500	Light west wind, clear.
10	54 "	32.0	3 "	4,500	Strong west wind, clear.
11	49 "	21.0	3 "	4,500	Light north wind, good logarithmic curve.
12	60 "	11.0	3 "	4,800	ditto.
13	66 "	29.4	12 "	4,000	North wind, strong.
14	59 "	13.0	3 "	4,500	Still, clear.
15	61 "	34.0	13 "	>5,000	Clear, west wind, light.
16	36 "	26.0	4½ "	4,500	Cold and foggy, west wind strong.
17	4 hrs.	56.0	13 "	1,500	Cold and foggy, barom. low and falling.
18	44 min.	3.8	3 "	5,000	Same, except barom. rising rapidly.
19	45 "	2.7	3 "	5,000	ditto.
20	51 "	11.6	3 "	1,700	Cold, damp, still.
21	43 "	7.2	3 "	4,000	ditto.
22	90 "	?	12 "		Exposure with A.C. 2,200 volts.
23	56 "	9.2	18 "	>5,000	North wind, very dry.
24	36 "	7.5	12 "	>5,000	ditto.
25	5¾ hrs.	70.3	22 "	500	ditto.
26	50 min.	11.7	4¾ "	4,000	Just after north wind.

$\lambda = .063$ and t is expressed in hours. This gives the decay of the part of the activity due to the thorium deposit. From the standard curve for the decay of radium A, B and C, in an equilibrium mixture the decay of the part of the curve due to radium active deposit was then determined, making the calculated and experimental values agree at the thirty-minute point. The thorium and radium excited activity decay curves were then added together and in prac-

TABLE II.

		0	½	1	1½	2	3	6	11	18	24
1	I	182	110	64	35	15	2				
	II	6	5.5	5.5	5	5	5				
	III	188	115.5	69.5	40	20	7				
	IV	188	116	61	33	16	6				
2	I	185	113	70	37	21	2.5				
	II	5.5	5	5	4.7	4.5	4				
	III	190.5	118	75	41.7	25.5	6.5				
	IV	190.5	118	70	41	24	7				
3	I	260	160	90	46	30					
	II	10	9.5	9	8.5	8					
	III	270	169.5	99	54.5	38					
	IV	270	170	100	56	37					
4	I	160	88	40	22	6					
	II	10	9.8	9.6	9.3	9					
	III	170	97.8	49.6	31.3	15					
	IV	170	94	47	27	14					
5	I	164	117	69	31	15	6	?	0	0	0
	II	24	23	22	21.5	21	19	17	12.3	6.7	5.0
	III	188	140	91	52.5	36	25	=	12.3	6.7	5.0
	IV	188	130	83	58.0	40	28	20	12.3	6.7	4.0
6	I	150	90	53	30	12	7	?	0	0	0
	II	22	21	21	20	20	19	15	11	6	4.8
	III	172	111	74	50	32	26	=	11	6	4.8
	IV	172	110	71	54	40	28	16	11	6	4.8
7	I	138	67	41	23	12	6				
	II	39	38	38	37	36	35				
	III	177	105	79	60	48	41				
	IV	177	105	77	58	48	41				
8	I	190	128	80	48	24					
	II	46	44	43	42	40					
	III	236	172	123	90	64					
	IV	236	168	124	90	64					
9	I	85	56	36	20	10					
	II	73	71	68	66	64					
	III	158	127	104	86	74					
	IV	158	127	108	91	73					
10	I	127	61	39	27	11					
	II	46	44	43	42	41					
	III	173	105	82	69	52					
	IV	173	106	82	61	51					
11	I	153	87	54	30	15					
	II	40	39	38	37	36					
	III	193	126	92	67	51					
	IV	193	127	89	66	50					

TABLE II. — *Continued.*

		0	½	1	1½	2	3	6	11	18	24
12	I	53.4	37	23	13	7	2				
	II	6.6	6	6	6	5.5	5.5				
	III	60	43	29	19	12.5	7.5				
	IV	60	44	30	19	13	8				
13	I	110	66	40	21	11	4	?	0	0	0
	II	46	45	43	42	41	39	32	24	14	11
	III	156	111	83	63	52	43	=	24	14	11
	IV	156	109	82	64	54	45	32	23	14	9
14	I	261	172	126	63	39	8				
	II	39	38	36	35	34	30				
	III	300	210	162	98	73	38				
	IV	300	210	148	100	73	36				
15	I	173	93	55	32	17	4				
	II	97	94	91	88	86	80				
	III	280	187	146	120	103	84				
	IV	280	187	141	117	103	85				
16	I	210	82	50	30	14	5			0	
	II	74	72	69	67	65	61			21	
	III	284	154	119	97	79	66			21	
	IV	283	160	116	94	80	70			28	
17	I	43	27	18	11	6	2	?	0	0	0
	II	55	54	53	50	48	45	37	28	18	11
	III	98	81	71	61	54	47	=	28	18	11
	IV	98	81	71	63	56	47	37	28	20	14
18	I	190	111	65.5	32	17.5	8				
	II	7.5	7	6.5	6	5.5	5				
	III	197.5	118	72	38	23	13				
	IV	197	118	68	38	24	13				
19	I	214	136	76.5	59	35	15				
	II	6	6	5.5	5	5	5				
	III	220	142	82	64	40	20				
	IV	220	142	82	64	40	20				
20	I	152	88	54	30	16	6				
	II	20	20	18	18	16	16				
	III	172	108	72	48	32	22				
	IV	172	112	82	54	36	25				
21	I	154	90	56	34	19	4				
	II	12	12	11	11	11	10				
	III	166	102	67	45	30	14				
	IV	166	102	66	44	31	15				
23	I	99	57	36	18	4	2	?	0		
	II	10	10	10	9	9	9	8	5		
	III	109	67	46	27	13	11	=	5		
	IV	109	67	43	26	13	11	8	6		

TABLE II. — *Continued.*

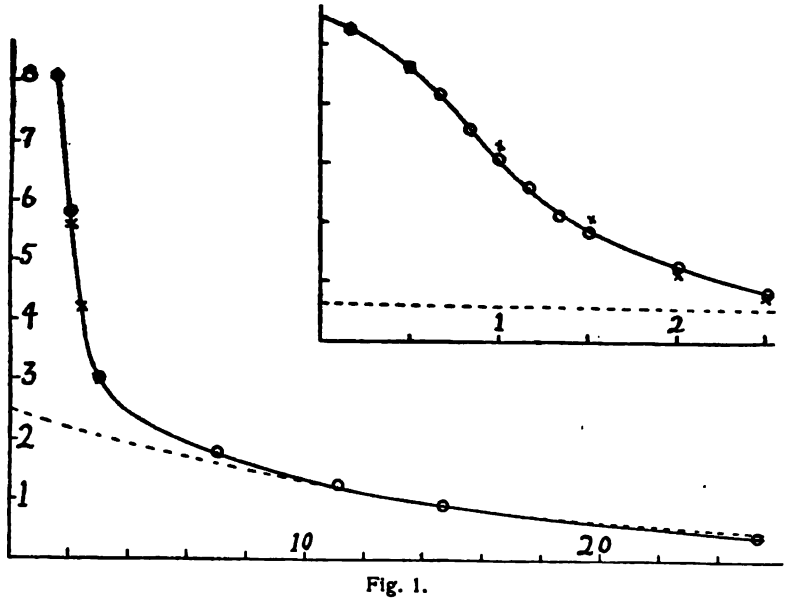
		0	½	1	1½	2	3	6	11	18	24
24	I	185	83	55	31	14	3	?	0		
	II	15	14	14	13	12	12	9	7.5		
	III	200	97	69	44	26	15	=	7.5		
	IV	200	97	62	40	26	14	9	7.5		
25	I	64	45	33	22	8	2	?	0		
	II	151	145	135	132	130	124	103	75		
	III	215	190	168	154	138	126	=	75		
	IV	215	190	170	155	142	127	103	75		
26	I	468	280	174	101	47	14	0	0		
	II	62	60	56	53	51	48	40	32		
	III	530	340	230	154	98	62	40	32		
	IV	530	340	230	154	98	60	40	32		

tically all cases the agreement with the observed values is closer than the limit of experimental error.

Table II. shows the agreement between the observed and calculated results. The number at the head of each column refers to the time, in hours, from the end of the exposure. The number at the left of each set of four rows refers to the number of the observation. The Roman numerals are as follows in each case: I. is the ionization due to the radium products; II. is that due to the thorium products; III. is the sum of I. and II. and gives the calculated activity; IV. is the observed activity. A few curves are given for illustration (see Figs. 1–8, illustrating observations 5, 6, 11, 12, 17, 18, 19 and 21 respectively).

The percentage of thorium excited activity varies between very wide limits (see Table I.), the minimum obtained being 1.8, the maximum 70.3 per cent. of the total activity. The following general statements may be made regarding the conditions governing the percentage of thorium excited activity.

1. A much larger percentage of thorium excited activity collects on a wire exposed at a low potential than on one exposed at a high potential in the open air. This point has been thoroughly tested. Rows 20 and 21 of Table I. are a fairly good illustration of this although here the potential was not varied greatly. The results shown in row 20 were obtained when the potential was — 1,700


8
7
6
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20
1
2

Fig. 1.

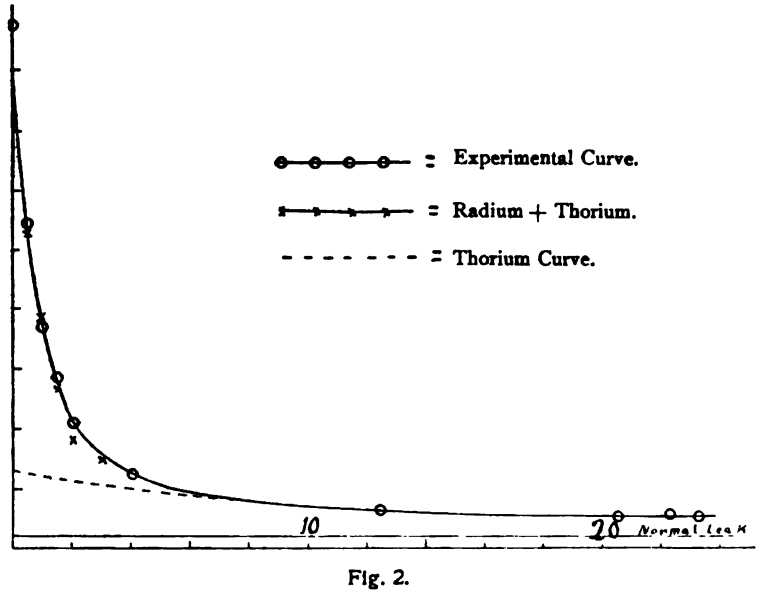

= Experimental Curve.
= Radium + Thorium.
= Thorium Curve.
10
20
Normal Leak

Fig. 2.

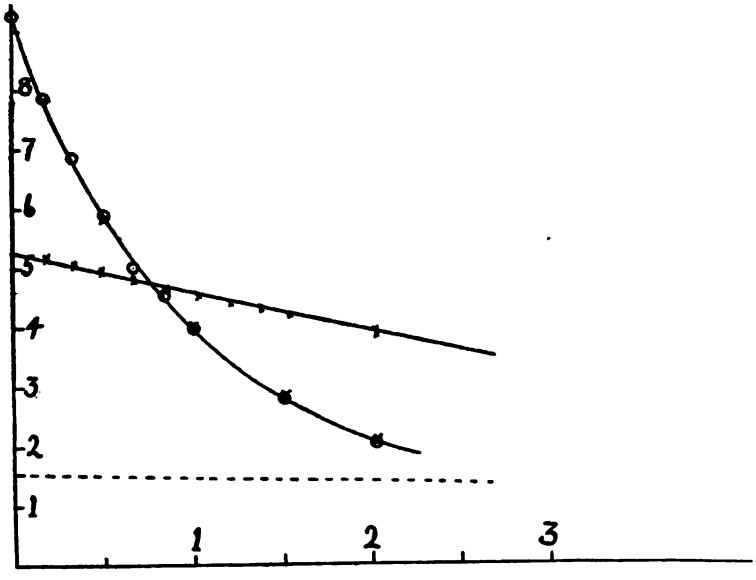


Fig. 3.

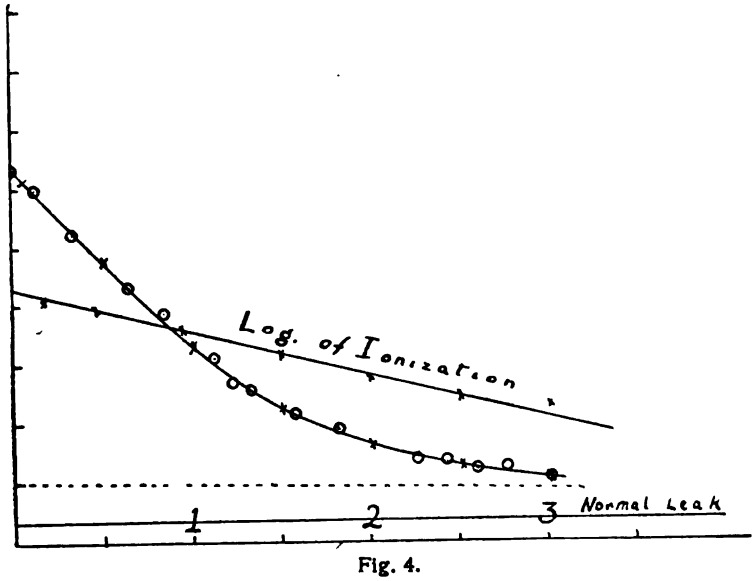
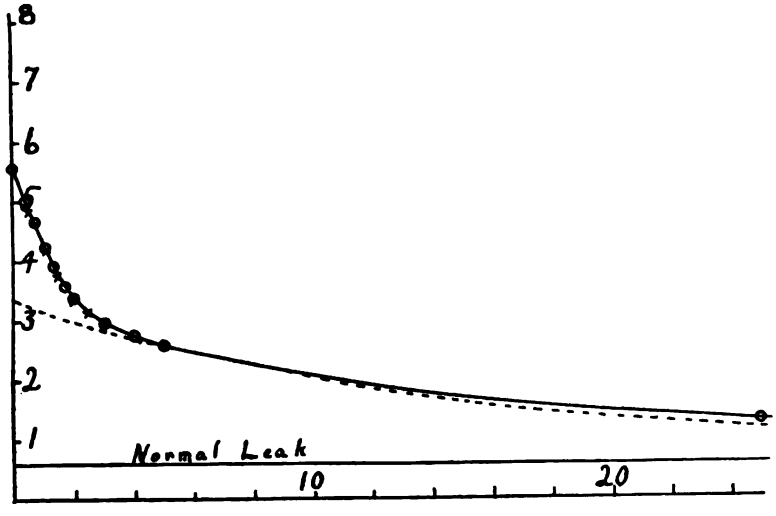


Fig. 4.


8
7
6
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Normal Leak
10
20

Fig. 5.

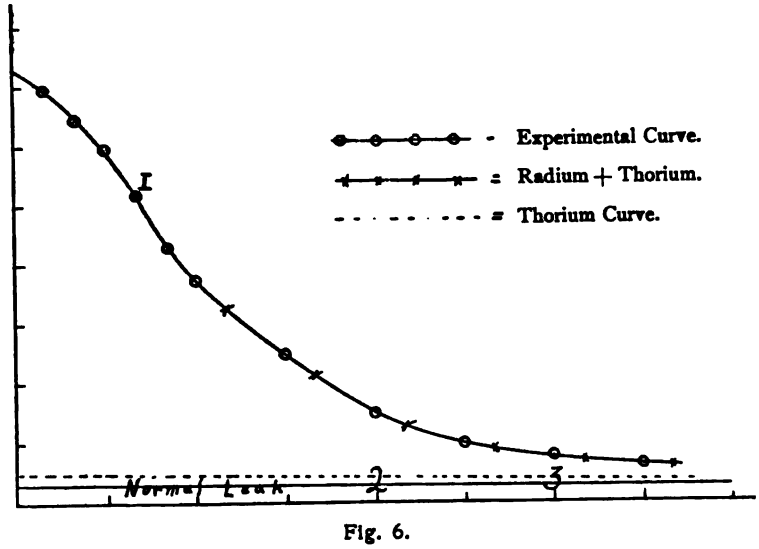
Experimental Curve.
= Radium + Thorium.
= Thorium Curve.
I
Normal Leak
2
3

Fig. 6.

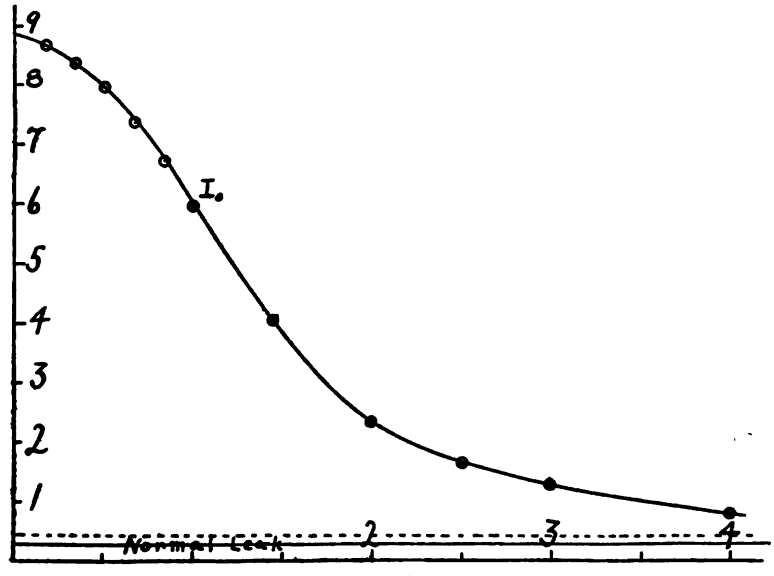

Fig. 7.

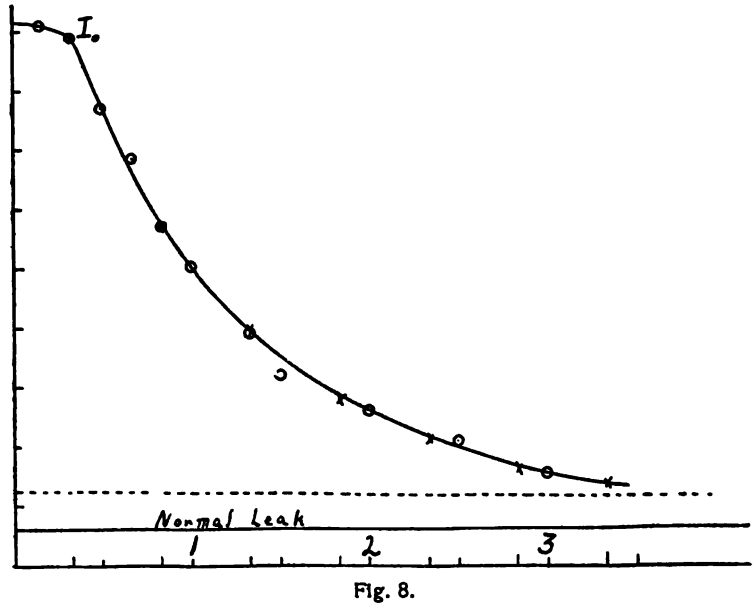

Fig. 8.

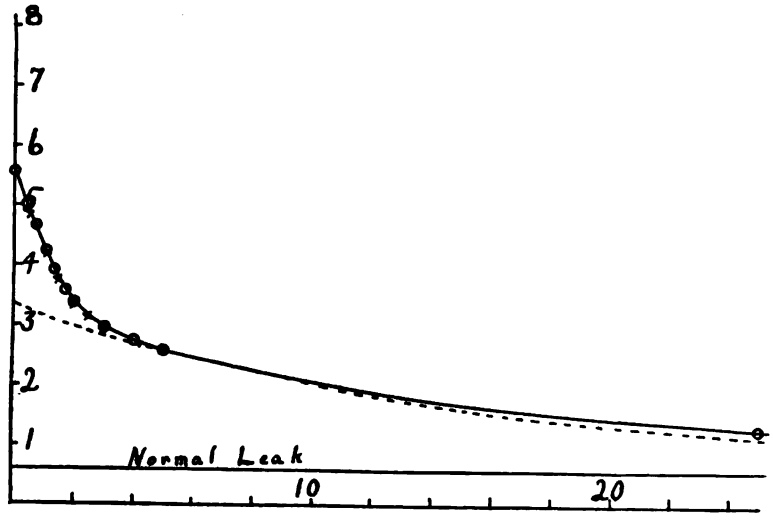
8
7
6
5
4
3
2
1
Normal Leak
10
20

Fig. 5.

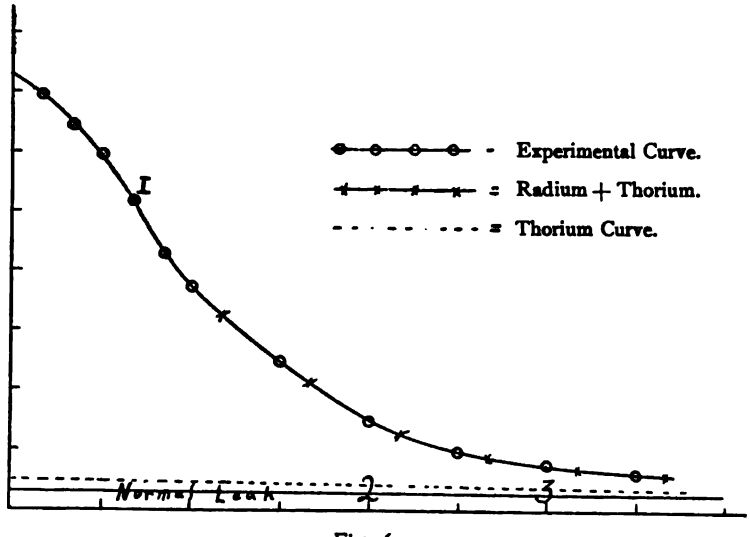
Experimental Curve.
= Radium + Thorium.
= Thorium Curve.
I
Normal Leak
2
3

Fig. 6.

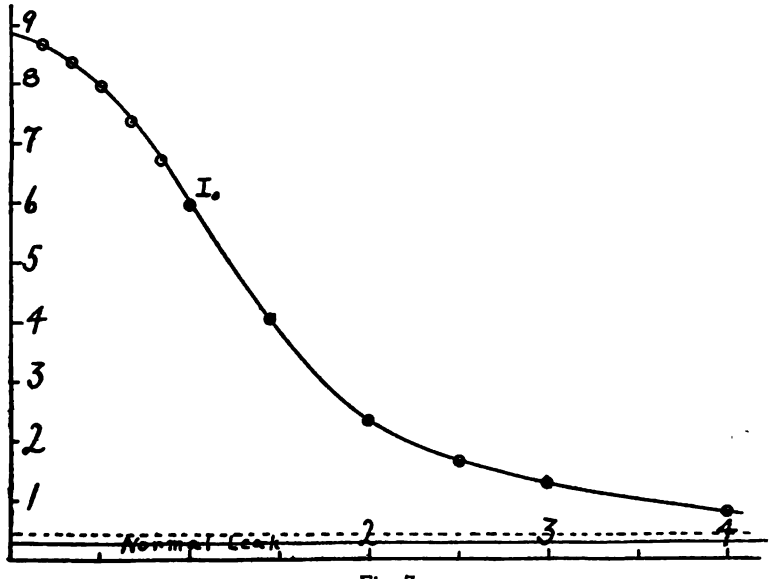

Fig. 7.

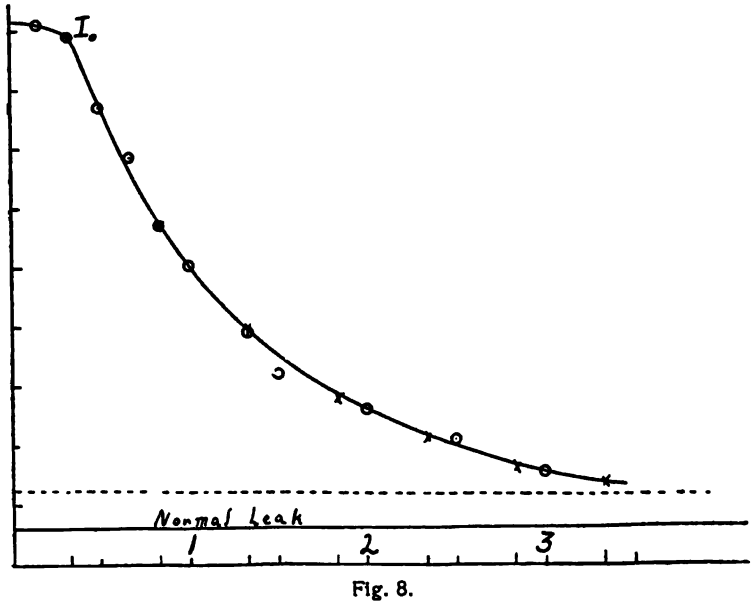

Fig. 8.

volts, in row 21 when it was — 4,000 volts. Atmospheric conditions were identical in the two cases and the two exposures occupied a total time of only seven and a half hours. Row 20 shows 11.6 per cent., row 21, 7.2 per cent. of thorium excited activity. Row 17 shows an unusually high percentage. The low potential, — 1,500 volts is partially responsible for this although here other conditions combine to cause the high percentage (see below). Row 25 which shows the highest observed percentage, 70.3 per cent., of thorium deposit is perhaps the most striking example. The observations shown in this row were taken under conditions very similar to those of row 23 and 24, *i. e.*, during a north wind, when a low percentage would be expected (see below). The potential was very low, being only — 500 volts. The time of exposure was long but not greatly longer than that of the wire in observation No. 23.

This fact contains the explanation of the very large percentages, from 50 to 70 per cent., of thorium excited activity obtained at Rome by Blanc,[1] who used a potential of only — 500 volts. It has long been known that this is not a sufficiently high potential to collect the maximum amount of active deposit on a wire exposed in the open air. These percentages, it is true, were obtained with very long exposures, and it takes a little over three days for the thorium active deposit to reach 99 per cent. of its final value. It is a very simple matter however to calculate the final per cent. of thorium deposit, which would have been collected on the wire had equilibrium been reached, from the time of exposure and the observed percentage. Moreover this is a much more reliable method as it is very unlikely that the percentage of thorium excited activity would not change during a three-day exposure and this would produce irregularities in the curve which would militate against accuracy. In Table III. the calculated percentage of thorium excited activity, had it reached an equilibrium is given under the heading per cent. Th. Corrected, and it is here conclusively shown that there is a very wide variation in the actual proportion of thorium excited activity to be collected from the atmosphere at different times, the minimum being 10.6 per cent., (row 24) and the maximum 76 per cent. (row 9). Sixteen out of the twenty-six values fall below the minimum set by

[1] G. A. Blanc, Phil. Mag., 13, p. 378, March, 1907.

TABLE III.

No. of Curve.	Hours of Exposure.	Total Activity.	Due to Ra.	Due to Th.	Ra Corrected.	Th Corrected.	Total Corrected.	Per Cent. Th Corrected.	Barom.
1	3½	188	182	6	190	30	220	13.6	Steady.
2	4	190.5	185	5.5	191	25	216	11.5	"
3	3	270	260	10	271	59	330	17.8	
4	3	170	160	10	167	59	226	26.5	Rising.
5	12¾	188	164	24	164	44	208	21.2	
6	14½	172	150	22	150	39	189	20.6	Falling.
7	3	177	138	39	144	230	374	61.5	"
8	4	236	190	46	196	209	405	51.6	
9	5	158	85	73	86	270	356	76	
10	3	173	127	46	133	270	403	67	
11	3	193	153	40	202	235	437	54	Steady.
12	3	60	53.4	6.6	63	39	102	38.2	"
13	12	156	110	46	156	87	243]	35.6	"
14	3	300	261	39	272	230	502	46.0	"
15	13	280	173	97	173	173	346	50.0	"
16	4⅓	284	210	74	216	308	524	59.0	
17	13	98	43	55	43	97	140	69.3	Low and falling.
18	3	197	122	7.5	127	44	171	25.7	Rising rapidly
19	3	220	214	6	223	35	258	13.6	ditto.
20	3	172	152	20	158	118	276	42.8	Falling slightly.
21	3	166	154	12	160	70	230	30.2	Steady.
23	18	109	99	10	99	19	118	16.1	Steady.
24	12	200	185	15	185	22	207	10.6	Rising.
25	22	215	64	151	64	198	262	75.8	"
26	4⅓	265	234	31	238	120	358	33.6	Steady.

Blanc, viz., 50 per cent. I think that without doubt the lower value of the percentage of radium emanation given by Blanc, for Rome, compared with that by Dadourian[1] for New Haven is due to the fact that Blanc did not use sufficiently high potentials, rather than the difference in locality.

An explanation of the cause of the higher percentages of thorium excited activity with low potential is somewhat difficult owing to the imperfect state of our knowledge as to the actual mechanism of the collection of the active deposit on a negatively charged wire; *i. e.*,

[1] H. M. Dadourian, Amer. Jour. Sc., 25, p. 335, April, 1908.

why, when the neutral emanation expels a positively charged α-particle it leaves behind a positive instead of a negative charge on the radium A particle. There must be, it would seem, a different rate of diffusion because of which most of the thorium A particles are drawn to the wire for a low potential while it requires a high potential to exhaust the surrounding air of its radium transformation products.

2. The percentage of thorium excited activity is very sensitive to barometric change. Observations 17, 18 and 19, represented by Figs. 5, 6 and 7 furnish a good illustration of this. The potential is, it is true, lower for Fig. 5, but the extreme change from 69.3 to 13.6 per cent. (see Table III.) of thorium deposit is not all accounted for by this difference. Observations 17 were taken just at the end of a long continued barometric fall, 18 and 19 immediately afterward when the barometer was rising rapidly. The percentage decreases from observations 18 to 19, although one was taken immediately after the other and conditions were exactly similar. This plainly shows the decrease of thorium excited activity during a rapid rise in the barometer. It is to be expected if we suppose the emanation to diffuse upward through the soil. An increase of pressure would tend to prevent the escape of the emanation into the air and as its period is so short, 54 seconds, the supply of thorium A would be cut off at once and the only active products which it would be possible to collect would be the thorium A, B and C already present in the atmosphere. These would of course be decreasing all the time. The radium emanation, half-period 3.7 days, on the other hand, would persist much longer and keep up the supply of radium A. It is thus clear why the percentage of thorium active deposit is decreased by a sudden rise in the barometric pressure.

3. Another fact of interest for comparison is the following: D. Pacini,[1] finds for a wire exposed over the Gulf of Liguria, for nine hours, that practically all the activity is due to radium, with scarcely a trace due to thorium. At Berkeley the west wind sweeps directly in through the Golden Gate, over the Bay of San Francisco and over about 2.4 miles of land to the Physics building, yet the percentages of thorium excited activity are much larger for this wind than for

[1] D. Pacini, N. Cimento, 15, p. 24, January, 1908.

any other. The average of the equilibrium values of the per cent. of thorium excited activity, from Table III., for exposures 7, 8, 9, 10, 15 and 16, which were made during periods of west wind, is 60.8 per cent. The average of all other percentages except those from north wind exposures 37.9 per cent. The average of all percentages except for west wind exposures is 31.7 per cent. These percentages prove that either the air over the Pacific Ocean off San Francisco, does contain thorium emanation or that the West Berkeley tidal-marshes have an unusually high percentage of thorium salts. Opportunity has not yet presented itself to test the two possibilities.

The initial activity on the wire varied between very wide limits, the maximum being as much as seventy times the minimum. The greatest amount of active material was collected during a hot, dry, north by east wind. This wind is known in Berkeley as the "north wind" and is the only strong land wind. It attains a velocity of 20 or 30 miles an hour at times. There is a large area covered by volcanic springs, where Shasta, Tehama and Plumas counties, California, come together, about 180 miles from Berkeley. The north wind crosses this area and follows the course of the Sacramento River. It seems very probable that the increased activity, at such times, is due to radium emanation carried down from this region. This supposition is borne out by the fact that the greatest activity does not appear on the first day of the wind, which usually blows three days, and also by the fact that the increase in the activity is very largely due to radium products whereas there is not a corresponding increase in the activity due to thorium products. The short life of the thorium emanation would account for this latter. Table III. does not give the actual initial values of the activity on the wires as it is more convenient to have comparable numbers for the purposes of plotting, etc., but it will be seen that the sets of observations 1 to 6, 23 and 24 show a low percentage of thorium excited activity. These observations were taken during periods of north wind. The results shown by curve 25 apparently present an exception to this as they also were taken at the time of a north wind but here the high percentage is due to the low potential as explained above. The low percentage shown in rows 18 and 19 is also explained above.

The north wind is accompanied by very disagreeable physiological effects and it has been suggested that these may be due to the abnormally large amounts of radium emanation in the air. Experiments on the effect of radium emanation on animal life would be comparatively easy to try and might yield some interesting results.

The north wind is also accompanied by a rise in temperature and low relative humidity. Ordinarily, at Berkeley, the relative humidity varies from 80 to 90 per cent., but at these times it may fall as low as 50 per cent.

The south wind is also a land wind, but is not accompanied by nearly such large amounts of active material. The relative humidity is so high that insulation is maintained with extreme difficulty and measurements hard to make.

The lowest activity accompanies the wind from the west, *i. e.*, from the ocean. Here also the relative humidity is high. The experiments of Elster and Geitel [1] showing that the greatest amount of active deposit was collected during a fog are not borne out at Berkeley. The fog here is not a land fog but is blown in from the ocean and is accompanied by a very low activity. The greatest amount of active material is collected when the relative humidity is low, *i. e.*, when there is a north wind as stated above. Attempts to discover a direct relation between humidity and amount of active deposit have not been successful as might be predicted since too many disturbing conditions enter.

The variation in temperature is so small at Berkeley that no effects due to it have been observed.

The statement is very generally made that "the height of the barometer was found to exert a very marked influence on the amount of excited activity to be derived from the air."[2] This appears to need correction in its statement though not in its interpretation. It is not the actual atmospheric pressure which has an effect on the amount of active material but the history of the barometer for a few hours previous to and during the exposure. It has already been shown that a sudden rise in the barometer will cause a change in the relative amounts of thorium and radium deposits but not very

[1] Elster and Geitel, Phys. Zeitschr., 4, p. 522, 1903.
[2] Rutherford, Radioactivity, second edition, p. 518.

much change in the total amount of material collected. A long continued low barometer usually causes an increase in the amount of active material collected but the north wind, which, as has been said, brings the greatest amount, is almost always accompanied by a steady, high barometer.

It appeared from the earlier experiments that an uncharged wire did not collect a measurable amount of active deposit. This was true for a wire suspended near the Physics building and running parallel to its walls. In the later experiments a wire was stretched across the roadway, near the building, and it was found that considerable quantities of dust were collected. The uncharged wire was therefore tested again after an exposure of some 48 hours and a slight activity detected. This result again agrees with the work of Allen (*loc. cit.*) on the Radioactivity of a Smoke-Laden Atmosphere. It appears that the active particles collect on the dust or smoke particles which in turn collect on the wire. No such large amounts as Allen speaks of were at any time collected. A positively charged wire showed no signs of activity after a twelve-hour exposure. A wire connected to one terminal of a 2,200 volt alternating current circuit while the other terminal was earthed showed more activity than an uncharged wire and less than one charged negatively to the same potential.

In order to determine whether the fact that the wire is charged during an exposure has any effect on the period of transformation of the particles which collect on it the following experiments were tried. Two wires were exposed, side by side, charged to the same negative potential for five hours. At the end of this time they were brought in and the activity of one of them tested. The other was coiled in a convenient shape and hung in a bell-jar of small capacity, where it was maintained at the same negative potential it had when exposed. After two and a half hours its activity was tested and it was found to continue the curve of decay from the earlier wire without discontinuity. The experiment was repeated maintaining the wire when in the bell-jar positively charged, with the same result as for the negative charge. This shows that the charge on the wire does not have any effect on the period of transformation. The result might again have been predicted as, accord-

ing to the theory of radioactive transformations, no force of finite magnitude can alter the period of transformation.

Curves of the shape shown in Figs. 6 and 7, consisting at first of a convex upward part, are very similar to those called class 2 by S. J. Allen (*loc. cit.*). I have found that a curve of this type almost always results from an exposure made while the barometer is rising, and seldom from an exposure made under any other conditions. It is to be expected that a change in the barometric height would cause a change in the supply of emanation and hence a change in the equilibrium value of the mixture of products and hence an irregularity in the shape of the curve.

OBSERVATIONS MADE IN DENVER.

During June and July, 1906, some measurements of the active deposit obtained from the atmosphere were made in Denver, Colorado, and as I am not aware of any published results from this locality they are given here.

The exposed wire was of copper, No. 18 B. and S. gauge about 12 meters in length and three meters from the ground. The potential was maintained by means of a water-dropper. After an exposure the wire was wound in the form of a helix and introduced into an aluminium-leaf electroscope, where the rate of decay was followed by the method 1*a*, described above.

The results do not differ essentially from those obtained at Berkeley except as to the effect of the direction of the wind. The greatest amount of active deposit was collected at the time of a high northwest wind but as this was the only strong wind during the two months over which the observations were extended, the direction cannot be regarded as having any very great importance. The amount of active material collected does not differ appreciably at the two places. Percentages of thorium excited activity and the half-period vary between about the same limits.

The observations were brought to an end by the maturing of the seed-pods on the cotton-wood trees. The blowing of the cotton made insulation a matter of extreme difficulty.

Of the exposures made, nine yielded useful results. The tables corresponding to Tables I. and III. follow.

TABLE D–I.

No. of Curve.	Half-Period.	Per cent. of Th Initially.	Time of Exposure.	Potential.	Remarks.
D–1	90 min.		3 hrs.	4,000	High west wind.
D–2	30 "		3 "	4,500	ditto. Short period due to Ra. A.
D–3	60 "	50	15 "	4,500	Very high northwest wind.
D–4	53 "	27	4 "	4,500	Clear, still.
D–5	55 "	13.7	4 "	3,000	ditto.
D–6	48 "	8.0	3½ "	4,250	ditto.
D–7	45 "	4.0	4 "	4,500	ditto.
D–8	50 "	2.5	3 "	3,700	Cloudy, still.
D–9	33 "	15.0	3¼ "	4,750	Light east wind, clear, short period due to Ra. A.

TABLE D–III.

No. of Curve.	Hours of Exposure.	Total Activity.	Due to Ra.	Due to Th.	Ra Correc.	Th Correc.	Total Correc.	Per cent. Th Corrected.
D–3	15 hrs.	540	270	270	270	440	710	62.0
D–4	4 "	130	95	35	98	159	257	62.0
D–5	4 "	127	109.6	17.4	113	79	192	41.0
D–6	3½ "	180	165.6	14.4	173	72	245	29.4
D–7	4 "	400	384	16	396	73	469	15.6
D–8	3 "	440	429	11	452	65	517	12.5
D–9	3¼ "	380	323	57	334	317	651	48.7

RANGE OF THE α-PARTICLES FROM THE ACTIVE DEPOSIT AND FROM RADIUM B.

It seemed desirable as a further proof that the active deposit collected on a negatively charged wire exposed in the open air consists of radium and thorium transformation products, to test the range of the α-particles. It was thought that the method proposed by Bragg [1] for a feebly active product might be used, for, as has been shown, wires can be obtained with a very low percentage of thorium excited activity and this amount might be calculated and the proper correction made. The method measures the range in terms of the stopping power of a screen of variable thickness covering the active substance. A wire was therefore exposed, the active deposit rubbed off with a piece of absorbent cotton moistened with ammonium hydroxide, a treatment which will further reduce the

[1] W. H. Bragg, Phil. Mag., 11, p. 754, June, 1906.

percentage of thorium active deposit as this deposit is less soluble in ammonium hydroxide than that of radium. The cotton was then incinerated, the residue spread in as thin a layer as possible and its activity tested, first when the ionization chamber was directly exposed to its influence and then as very thin sheets of aluminium foil were placed over it. Readings were frequently taken with the naked deposit to obtain data from which the decay curve could be plotted. The time of all readings was recorded. The result obtained was very unexpected. Fig. 9 plainly shows the presence of a radiation which

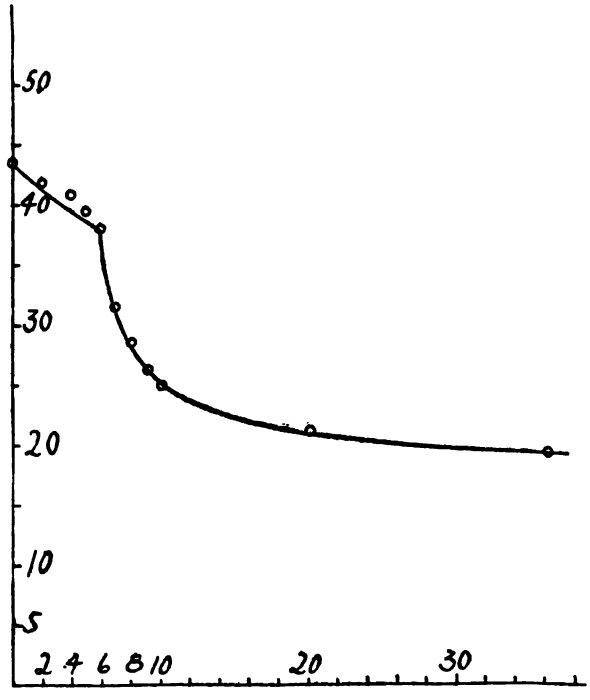

Fig. 9.

is completely absorbed by from 6 to 10 layers of aluminium foil. It is evident at once from the curve that agreement among separate calculations of the full range cannot be expected by Bragg's method. The characteristic shown in the tables given by him, *i. e.*, an increase in the value of the product ρd, *i. e.*, density times thickness, for full range, as the thickness of the foil was increased was shown to a very marked degree. Calculations for the range are all smaller than

would be expected and the calculation from the foil of greatest thickness gives a little less than one half the range of radium C.

The recent work of S. J. Allen[1] explains this value. He has tested the range of the α-rays from the active deposit obtained from the atmosphere and has shown that "where the rays are allowed to emerge in all directions to the normal the position (*of maximum ionization*) is much less than the range of the particle, being roughly one half." The above result is in agreement with this statement by Allen.

The product, ρd, of the foil used to obtain the data for Fig. 9, was 6.47×10^{-5} for each sheet. Using the value of the stopping power of aluminium foil, in terms of air, given by Bragg, viz., foil for which $\rho d = .00329$ is equivalent to 2.30 cm. of air, calculation shows that the easily absorbed radiation would be completely stopped by between 2.7 and 4.5 mm. of air. The values of the stopping power of aluminium foil obtained by S. J. Allen (*loc. cit.*) agree very well with that of Bragg, and show that it is about the same for rays from radium C, thorium and uranium. It is to be seen from Fig. 9 that nearly half of the ionization produced by the naked deposit is due to this easily absorbed radiation. The curve has been corrected for the natural decay of the deposit and the ionization chamber was made deep enough to allow the α-particles from radium C and thorium C to run their full course.

It was of course desirable to determine which of the several components of the active deposit gives off this easily absorbed radiation. To do this, first, a wire was exposed for a long time in the open air in order to obtain a large amount of thorium active deposit. This deposit was allowed to stand for six hours before any tests were made, during which time all the radium products transformed into the inactive radium D. The data for an absorption curve were then obtained by adding layers of aluminium foil and taking readings of the ionization. The absorption curve, corrected for the natural decay, showed no trace of the easily absorbed radiation. The radiation, then, cannot be due to thorium A nor any of its successive transformation products. This result was expected as Fig. 9 shows such a large part of the total ionization to be cut off by the first ten sheets of foil.

[1] S. J. Allen, PHYS. REV., XXVII., p. 294, October, 1908.

A wire was next exposed to radium emanation and the absorption curve obtained as before. The characteristic inflection reappeared. It is not quite so marked as in the case of the atmospheric curve but it is evident that part, if not all, of the easily absorbed radiation comes from radium excited activity.

Four curves were then obtained which were so timed that the inflection point should come at 30, 50, 60 and 140 minutes from the time of discharge of the wire. The point of inflection is most marked in the first, less so in the second, still less in the third and does not appear at all in the fourth. Radium A has all transformed before the beginning of the observations, *i. e.*, in about ten minutes. The activity after about two hours is almost entirely due to radium C. The easily absorbed radiation cannot then be due to either of these and must therefore be due to radium B.

An effort was then made to measure directly the range of this radiation. Two methods of attack at once present themselves. The slit or bundle of tubes necessary to get rid of all except the normal rays may be dispensed with, since the distances used are so very small. Or the pressure of the gas in which the α-rays travel may be reduced to about one tenth atmosphere, when the distances will become large enough for the usual method. The first method was used for two reasons. First, apparatus was at hand with only slight alteration and, second, the radium bromide at present available is so weak as to make it desirable to utilize all the radiation possible. The results give justification for the first method. The apparatus is shown in section in Fig. 10 and does not differ from the ordinary form, used for these measurements, except in two points. First, as has been said, the bundle of tubes limiting the rays which reach the ion-trap to those shot off normally, was dispensed with. Secondly, a sheet of very thin aluminium foil (*B–C*, Fig. 10) was stretched tightly just below the plate *A*, connected with the electrometer. This sheet of foil was equivalent to about 0.5 mm. of air. It prevents the

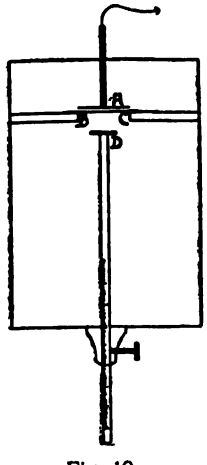

Fig. 10.

diffusion of ions from below and at the same time lets through more α-particles than a wire gauze. The plate A was set as close to the aluminium foil as possible without increasing the capacity of the system so much as to cause a large decrease in sensitiveness. The final distance used was about 7 mm. The plate was about 8

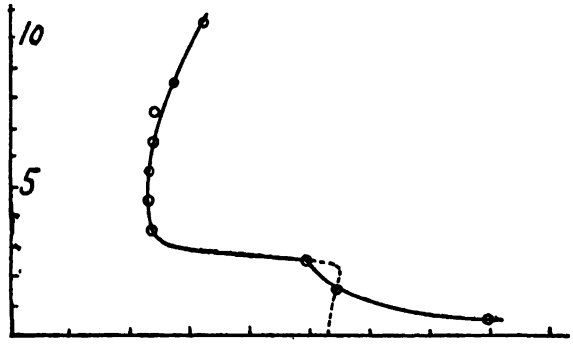

Fig. 11.

cm. in diameter. It is necessary to stretch the aluminium foil tightly as otherwise the attraction between the charged surfaces causes an alteration of the position of the aluminium foil.

The experimental curves obtained, as the distance of the wire, placed on the Table D, below the aluminium foil, was altered, are

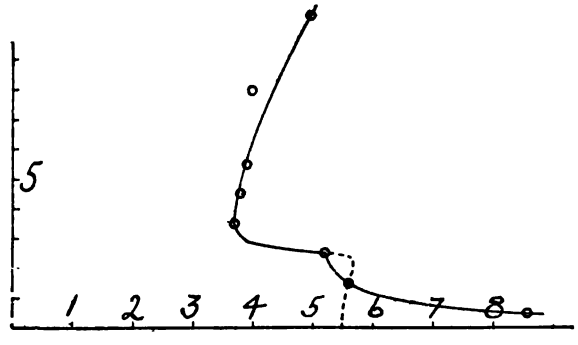

Fig. 12.

of the characteristic shape of those showing the range of α-particles, except at the very lower part (see Figs. 11 and 12). The cause of the departure from the usual, dotted form, is easily explained.

Theory requires that the ion-trap shall be shallow in comparison with the range of the α-particles. This is realized for the upper part of the curve where the α-particles from radium C produce the ionization. Hence the curve takes the usual form for this part, and for the part where the short range particles just begin to reach the ion-trap. As the distance is further diminished however the short range particles produce more and more ionization. When the wire touches the aluminium foil they run their full range and hence produce the greatest ionization. The figures show that the easily absorbed radiation has a range between 2.6 and 3.0 mm. in air under normal conditions. The curves are plotted from the following data, corrected for the natural decay of the active material, and for the absorption by the thin sheet of aluminium foil.

Distance in mm.	0.5	1.5	2.5	3.5	4.5	5.5	6.5	7.5	8.0	8.5	10.5
Ionization Fig. 11.	800	545	495	267	260	263	290	290		325	375
Ionization Fig. 12.	860	560	520	360	370	380			400		500

SUMMARY.

1. The half period of decay is not constant but varies from thirty minutes to five and a half hours.

2. Agreement between the experimental results and those calculated on the basis of a mixture of thorium and radium products is closer than the limit of experimental error.

3. The percentage of thorium excited activity for an equilibrium mixture varies between 10.6 and 76.0 per cent. of the total.

4. Percentages of thorium excited activity depend upon the following conditions: (a) A wire exposed at a low potential, — 500 volts, collects a much larger percentage than one exposed at a high potential, — 5,000 volts or over. (b) The percentage is very sensitive to barometric change. (c) The west wind is accompanied by the largest percentage, indicating that the air over the Pacific Ocean, off San Francisco, contains thorium emanation or that the tidal marshes over which the wind comes have an unusually high percentage of thorium salts.

5. The greatest amount of active material is collected on days when there is a north wind.

6. Variation in atmospheric pressure and not actual barometric height affects the amount collected.

7. Curves which have initially a convex upward part are obtained when an exposure is made while the barometer is rising.

8. The active deposit obtained at Denver, Colorado, does not differ appreciably from that obtained at Berkeley, California.

9. Results on the range of the α-particles from the active deposit are in agreement with those obtained by S. J. Allen for the long range rays.

10. The short range α-particles given off by the active deposit are due mainly if not entirely to radium B.

11. Nearly half the ionization produced by the deposit obtained from an open-air exposure is due to α-rays which have a range of between 2.7 and 3 mm. in air, under ordinary conditions of temperature and pressure.

In conclusion I wish to thank Professor E. P. Lewis for his unfailing interest and kindness during the course of my investigations.

UNIVERSITY OF CALIFORNIA,
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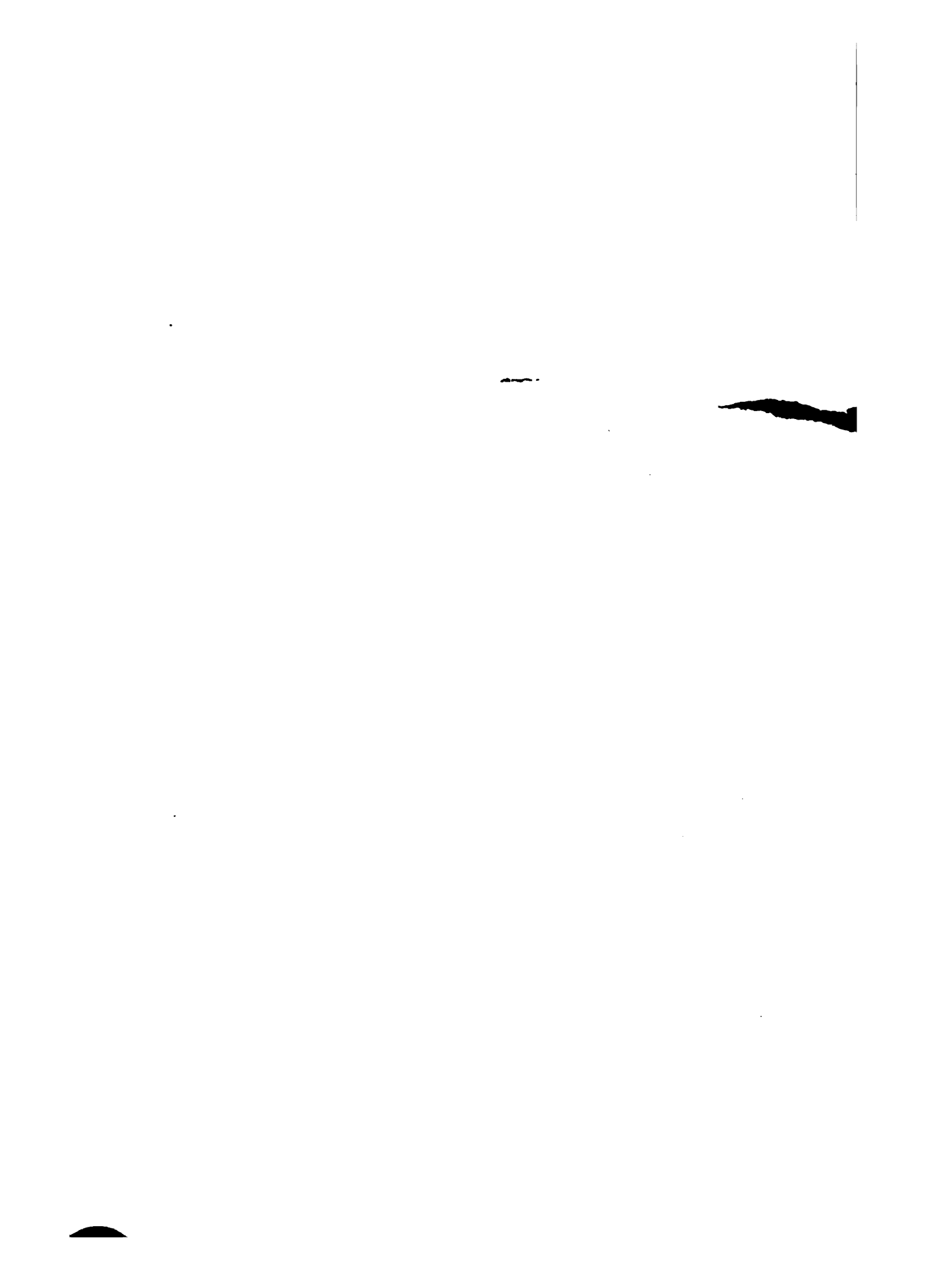

Harry